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(19)

(54) COEXTRUDED SHEET WITH PROPERTIES RESEMBLING A CROSS-LAMINATE AND METHOD OF PRODUCING SAID SHEET

(71) I, OLE-BENDT RASMUSSEN, a Danish subject, of 7, Topstykket, DK—3460, Birkerod, Denmark, do hereby declare the invention, for which I pray that a patent may be granted to me, and the method by which it is to be performed, to be particularly described in and by the following statement:—

The present invention relates to a method of manufacturing a laminated sheet and to apparatus for this.

Cross-laminates of uniaxially oriented films of crystalline polymers are known to exhibit a generally highly advantageous combination of different strength properties of which the most surprising has been the tear propagation strength (cf. US Patent No. 3,322,613) especially when the bonding between the layers is sufficiently weak that during tearing from an incission the layers will delaminate around the notch. As a result they split or flow in different directions and the notch effect is smoothed out, this being termed a "forking effect". Sheets of this kind are particularly useful for various heavy duty applications such as tarpaulin substitutes, cover sheets, heavy duty bags, and heavy duty wrapping film.

The most expedient method of producing a sheet of the above kind is described in British Patent Specification No. 816,607, and consists in strongly orienting the molecules of a tubular film in its longitudinal direction, helically cutting and unfolding it to a flat film with the orientation at bias (e.g. 45°C), and subsequently continuously laminating this film with a similarly produced flat film, while the respective directions of orientation are arranged in criss-crossing relationship.

It is known that, for a given thickness, the tear propagation resistance is pronouncedly increased by use of 3 layers with 3 different directions of orientation, e.g. obtained by laminating 1 longitudinally oriented film with 2 films which are oriented at bias as described above.

One drawback of the process described above (and the resultant product) is that it is practically impossible to produce really thin film, so that the economic advantage of producing a high strength but low weight film is not fully attained. In practice the lowest weight for each layer that can be achieved when spiral-cutting and laminating is about 30 g/M². Thus for a 2-layer laminate, the lower limit is about 60 g/M² while for a 3-layer laminate (which is mentioned above is necessary for proper utilization of the tear-stopping effects) it is about 90 g/M².

A second draw-back is the practical limitation in width caused by the rotation of heavy machine-parts and bobbins in connection with the spiral cutting. Generally the width is limited to about 1.5—2M.

A third draw-back relates to certain energy-absorption values for the cross-laminates. Relatively low energy-absorption has been found with regard to high-speed tearing (Elmendorf tear test) and for low and high speed tensile testing (TEA strength and Elmendorf impact strength). It appears that the very anisotropic character of the layers is disadvantageous. If for instance a 2-ply cross-laminate of this kind is drawn parallel to the direction of orientation in one of the layers, then the yield point and the elongation at break are in essence determined by that layer.

Earlier attempts to overcome the above-mentioned drawbacks, and to provide for a cheaper production process for a product with similar or analogous properties, is described in my British Patent Specification No. 1,261,397. In that specification a process is disclosed which produces a criss-crossing structure through a die with rotating parts, while forming in the same die a soft and weaker middle zone by coextrusion. The method comprises coextruding several concentric or almost concentric layers of crystalline polymer alternating with layers of a softer polymer,

and dividing the layers inside the die by means of teeth arranged in rows and fixed to the cylindrical die walls pointing from the concave wall surface inwardly, and from the convex wall surface outwardly. The die-parts are rotated in opposite directions and thereby the layers are divided according to lefthand helices near one and right-hand helices near the other sheet surface. The combing can either be carried through to the middle of the film or be limited to portions near the surfaces. The coextrusion of polymers before the combing zone is adapted to provide for a soft and weak middle-zone.

The film extruded by this method can be considered to be of unoriented material. However, the alternating stiff layers of a "first polymer" and soft layers of a "second polymer" divided to filaments in a linear pattern by the teeth imparts to each half-part of the sheet a tendency to split or flow in one direction, and since the linear patterns at the two surfaces criss-cross each other and a tendency to delaminate is provided, there is obtained a tear-stopping effect which is analogous to the "forking" effect in a true cross-laminate.

The above specification further proposes to biaxially stretch the laminate under such conditions that instead of yielding biaxially oriented layers the molecular orientation is generally unaxial in each layer, with the direction of orientation in different layer criss-crossing each other. In order to obtain such uniaxial orientation, the second material must be very prone to yield, e.g. because it is still molten or semi-molten while the first material is solid, and the filaments of the first material must be kept straightened out by biaxial strain.

Although the above method would in principle solve the problems of obtaining a lower thickness and higher width in cross laminates there were found some essential difficulties during the later technical developments. It was confirmed that the extrusion method was commercially feasible for manufacture of unoriented film with high tear propagation strength, but with a low impact strength due to the lack of orientation. However, essential drawbacks were found in connection with a subsequent biaxial stretching. As also indicated in the above specification, one must use a relatively great number of rows of the teeth in the extrusion die in order to obtain the fibre finess which is necessary for the stretching system.

This, however, made the maintenance of the die difficult and caused frequent "hang-up" of polymer lumps between the teeth. Further, the interaction between the teeth in one half-part of the die and those in the other half-part made it necessary either to use excessive amounts of soft middle layer material, or to limit the combing to two relatively thin surface zones of the sheet. Further, it was very difficult to establish and maintain the biaxial stretching conditions necessary for obtaining a generally uniaxial molecular orientation as described.

According to the invention a method of producing a laminate of at least two layers comprises the steps of rotating relative to one another in a circular coextrusion die having an exit slot at least two concentric tubular layers, each comprising a stream or an array of streams of molten polymer material, and simultaneously melt stretching each of said layers substantially in one direction, subsequently bonding the layers in the die immediately prior to their passage through the exit slot to form a laminate with the directions of melt stretching crossing each other and solidifying the laminate while maintaining the crossing melt stretched structure, the bonding in the solidified laminate being sufficiently weak to permit local delamination of the film upon tearing of the laminate.

Preferably the opposite sides of the exit slot through which the laminate is extruded are rotated relative to one another, since this results in the laminate being subjected to shear during extrusion.

The tubular layers are preferably formed of a dispersion of one polymer in a polymeric matrix, so as to produce by the melt stretching a grain of polymer along the direction of melt stretching, as described in more detail in my cognated copending applications 29807/74, 53644/74, 5971/75 and 5972/75 (Serial No. 1526722). Thus, as explained therein, a fibrillar grain structure having a predominant orientation of splittability after solidification of the laminate into a film may be obtained.

The two layers may be rotated, in the method of the invention, in different directions but with substantially the same angular speed.

One method of producing the desired weak bonding involves forming the layers of polymers that adhere poorly to each other. Another method involves coextruding between the layers a polymer for controlling the adhesive strength, this polymer for example being extruded in stripes or being otherwise interrupted. The adhesion controlling polymer may be an elastomer having poor adhesion for the polymer material or materials which form the tubular layers.

Each layer may be an array of streams, which will of course merge to form the layer, and the melt stretching may be performed by passing the material of this

array of streams through a row of partitions, for example as described below with reference to Figure 1.

The melt stretching may be performed by reducing the thickness of the molten tubular layer during extrusion.

Prior to uniting the layers with mutually crossing directions of stretching each layer may itself be formed from two or more other layers, namely by passing two or more tubular layers of different polymer material together through a common rotated die-part and coextruding them into a common chamber of the same rotated die-part, thereby forming a composite rotated tubular layer. Such a method is illustrated in Figure 2 of the accompanying drawings in which the coextrusion within the common rotated die-part takes place over a circular edge.

Preferably the laminate after solidification is biaxially stretched in the solid state in at least two separate steps each of which is essentially unidirectional. This stretching may be carried out substantially at room temperature. Generally it comprises stretching the sheet to a configuration of temporarily evenly distributed substantially longitudinal pleats by applying pressure along lines extending substantially in the longitudinal direction of the sheet, for example by passing in a nip between grooved rollers with the grooves parallel with or forming a small angle with the machine direction. This method of stretching the extrudate is described and claimed in Application No. 16900/78 (Serial No. 1526724), and, as described therein, gives transverse stretching. After the transverse stretching is completed the longitudinal stretching may be conducted, a substantial transverse contraction preferably being effected during the longitudinal stretching.

The polymer material for the tubular streams may consist mainly of polyolefin. Preferably at least one of the tubular streams consists mainly of crystallisable polypropylene or high density polyethylene. When a polymer for controlling bonding strength is coextruded between the tubular streams a suitable material is ethylene-propylene rubber.

For more details of methods of conducting the biaxial stretching, for polymer materials that may be used for the layers and for the properties of products that are obtainable by the method of the invention reference should be made to the aforementioned copending applications.

The invention includes not only the described method but also the apparatus for carrying it out when the weak bond is produced by coextrusion of a polymer between the layers. Such apparatus comprises a circular coextrusion die having an exit slot, means for feeding towards the slot at least two concentric tubular layers each comprising a stream or an array of streams of molten polymeric material, means for rotating the layers relative to one another in the die and for simultaneously melt stretching each layer substantially in one direction, means for coextruding between the said layers a polymer for controlling the adhesion strength and means for bonding the layers in the die, with their directions of melt stretching crossing each other, immediately prior to their passage from the exit slot. Preferably the means for rotating the layers in the die relative to one another comprise means for rotating the opposite sides of the exit slot relative to one another.

The invention will now be described in more detail with reference to the drawings, of which

Figure 1 shows a section through an extrusion die according to the invention;

Figure 2 shows in perspective view with displaced sections the principle of another extrusion die according to the invention with two counter-rotating exit slots and means to extrude two layers through each slot;

Figure 3 is a process-line of a preferred cold stretching method;

Figure 4 is a detail of the "grooved rollers" which perform the transverse stretching in uneven zones, called "striations";

Figure 5 is a schematical sketch, on an enlarged scale, of the pattern of the striations and the orientation therein, of a film cross-stretched according to the process-line of Figure 3;

Figure 6 is an enlarged cross-section of the film of Figure 5 as actually observed by microscopy. However for the sake of clarity, the thickness is shown on a scale twice that of the width.

The extrusion die shown in Figure 1 is an example of one that may be used, in which two polymer-in-polymer dispersions are extruded into a common collecting chamber through two rows of partitions, which are rotating in opposite directions. The two dispersion streams 1 and 2 are fed through inlet channels in the lower part of the die to annular channels 4 and 5 respectively in the two walls in the annular track 6, in which the two rings 7 and 8 are moved in opposite directions by driving means, e.g. by teeth and toothed wheels (not shown). The two rings 7 and 8 are supplied with rows of partitions 9 and 10 respectively, by which two rows of openings 11 and 12 are formed, through which the two dispersions are extruded into the collecting chamber 15, formed by the two parts 13 and 14, and

terminating in the exit slot 16. For the sake of simplicity the partitions 9 and 10 are shown as radially extending, but in reality they are placed at an angle to the radial direction to prevent the formation of die-lines in the extruded sheet. By the extrusion through the two rotating rings 7 and 8 the two dispersions each become attenuated and thereby acquire a fibrillar morphology and thus a direction of splittability, as discussed above. The two arrays of attenuated streams thereafter unite in the collecting chamber 15 to form a laminate with criss-crossing fibrous morphology. The thickness of this laminate is reduced by the passage through the exit slot 16 and further by a normal draw-down and blow process. Hereafter the film is stretched both in the longitudinal and the transverse direction at a relatively low temperature. Owing to the two different fibre directions the two half-parts of the film exhibit tendencies to split in different directions during tearing. The materials, from which the two half-parts are formed, are so selected that they adhere poorly to each other. The material thereby delaminates in a small area around the incision, from which tearing takes place, and this will smooth out the notch effect.

The die shown in Figure 2 consists of four main parts, viz. a fixed inlet part (17) for circular distribution of the polymers as explained below, a fixed bearing part (18), and supported here by the two rotating parts (19) and (20) which form one exit orifice (21). The polymers blends (A) and (B) are fed to the inlet part (17) where they are distributed in concentrical circular streams. (A) is extruded through the annular conduits (22) and (23), for which either one or two extruders may be used. (B) is extruded through the annular conduit (24). For even distribution, (22), (23) and (24) are supplied with distribution baffles or other distribution means (not shown).

For the sake of clarity, the bearings and sealings between the bearing part (18), the rotating part (19), and the rotating part (20), are not shown, neither are the drives for (19) and (20).

From the three anular conduits (22), (23) and (24) the polymer streams pass the bearing part (18) through three circular arrays of channels (25), (26) and (27), each communicating with an annular chamber (28), (29) and (30) respectively.

The two rotating parts (19) and (20) are preferably rotated at almost equal angular speed, but in different directions, as indicated by the arrows (31) and (32). Each rotary part in itself is a coextrusion die for two layers, one consisting of (A) and one of (B). For the sake of clarity, reference figures for explanation of the flow are shown only on part (20), but the flow through part (19) is similar. From the chamber (29) polymer blend (A) passes into the rotating part through channels (33), while polymer blend (B) from chamber (30) passes into the rotating part through channels (34). Inside the rotating part are two annular conduits (35) and (36) in communication with, respectively the channels (33) and (34), and separated from each other by a thin circular wall (37).

Having passed the edge of the wall (37), (A) and (B) merge together in an annular collecting chamber (38), which terminates in the exit orifice (21). By the passage through the annular conduit (35) and the collecting chamber (38), the thickness of the fluid sheet is strongly reduced whereby the material is attenuated.

The partitions between adjacent channels (33) and (34) respectively ought to be streamlined, as shown. For the sake of clarity they extend radially in the drawing, but in real fact they should be forming an angle with this direction to reduce the tendency of the forming of die-lines.

"Polymer A" is preferably a blend of two incompatible or semi-compatible polymers, while "Polymer B" is adapted to give the sheet a suitable tendency to delaminate. It may therefore, e.g. consist of an elastomer which is a poor adhesive for the two layers A, and may be extruded in stripes. However, if the channels 22 and 23 are fed with two different polymer blends, that are mutually incompatible, the polymer B may be an adhesive with a relative strong bond to the two polymer blends, and it must in that case be extruded in stripes or otherwise interrupted.

A preferred cold-stretching method is indicated by the process-line of Figure 3, in which section "Q" is the cross-stretching line and section "R" is the longitudinal stretching line. The system of rollers in section "Q" consists of driven nip rollers (71), driven grooved rollers (72) idle rollers (73) and rollers (74) having a longitudinal section resembling a banana. The banana rollers (74) after each step serve to draw out the pleats produced by the transverse stretching. Over the idle roller (75), the film (79) enters section "R", the longitudinal stretching line, where it is drawn through a water bath (76) serving to remove heat generated by the stretching and maintain a suitable stretching temperature e.g. at 20°—40°. Finally it is wound onto a bobbin (77).

The arrow (78) indicates the machine direction.

In Figure 4 a pair of driven grooved rollers (72) are shown in detail with the film (79) pressed and stretched between the teeth (80) of the rollers (72).

In Figure 5 the relative lengths of the

arrows in the striations I and II of the film (79) indicate the relative amounts of orientation achieved by the biaxial stretching method shown in Figure 3 and Figure 4.

In Figure 5 as well as in Figure 6 the numbers I and II indicates the striations, A and B respectively, discussed above, which generally have varying width and uneven character. Furthermore it should be noted, that the outer layers (81) and (82) of the film (79) are not always symmetrical with respect to the thin mid-layer (83). This asymmetry further serves to make a tear fork.

The following is an Example of the invention.

A series of sheets all based on polyolefin blends was produced by the extrusion die shown in Figure 2. The diameter of the exit slot (21) of the die was 130 mm and the width of the latter 1 mm. The greatest width of the collecting chamber (38) was 4mm, which means that the amount of attenuation during the passage through the collecting chamber toward the exit slot was smaller than is preferred. The extrusion temperature was about 240°C.

After the longitudinal cutting of the tubular film, the stretching was first carried out laterally using from 4 to 8 steps and thereafter longitudinally using from 2 to 4 steps. The composition, flat tube width (measure of blow ratio), stretching temperature, stretch ratio, and results appear from the table below. "Nov" stands for Novolene, a gas-phase polymerised polypropylene with relatively high contents of the atactic modification, "PE" stands for low density polyethylene, "EPR" stands for ethylene-propylene rubber, "SA 872" "7823" and "8623" are different types of polypropylene with minor contents of polymerised ethylene. EPR/PE stands for a 50:50 blend of ethylene-propylene rubber and low density polyethylene.

Run	Outer Layer Composition	Middle Layer Composition	Orientation Temp.	%	(cm) Tube Width	Tongue Tear MD	TD	Dart Impact (ft–lbs)
1	70% 7823, 20% PE, 10% EPR	PE–10%	Cold	100%	30	3.3	1.8	2.5
2	80% 7823, 20% EPR	PE–10%	Cold	100%	30	1.7	1.6	2.5
3	80% 7823, 20% EPR	PE–20%	Cold	100%	30	2.7	2.3	2.5
4	70% SA 872, 30% PE	PE–10%	Cold	50%	30	7.1	4.6	2.0
5	,,	PE–10%	Cold	100%	30	5.7	2.1	2.5
6	,,	PE–10%	Hot	50%	30	9.0	4.5	2.0
7	,,	PE–10%	Hot	100%	30	7.5	3.0	1.5
8	,,	PE–10%	Cold	50%	45	6.6	1.4	2.0
9	,,	PE–10%	Cold	100%	45	2.6	1.4	2.0
10	,,	PE–10%	Hot	50%	45	5.8	2.4	2.0
11	,,	PE–10%	Hot	100%	45	6.4	2.3	2.2
12	100% SA 872	PE–10%	Cold	100%	30	1.2	3.3	2.5
13	,,	PE–10%	Cold	100%	30	2.0	1.1	1.5
14	,,	EPR/PE–10%	Cold	100%	30	2.1	1.5	2.5
15	,,	EPR/PE–10%	Hot	50%	30	3.1	2.1	1.5
16	,,	EPR/PE–10%	Hot	100%	30	1.4	1.4	1.5
17	,,	EPR–10%	Cold	100%	30	5.6	2.8	2.0
18	80% 7823, 20% EPR	EPR–10%	Cold	100%	30	4.4	1.9	4.0
19	70% 7823, 20% PE, 10% EPR	EPR–10%	Cold	100%	30	4.5	2.9	3.0
20	70% SA 872, 30% PE	EPR–10%	Cold	50%	30	6.4	3.0	1.5
21	,,	EPR–10%	Cold	100%	30	3.9	2.1	3.0
22	,,	EPR–10%	Hot	50%	30	4.9	3.9	1.5

TABLE (Continued)

Run	Outer Layer Composition	Middle Layer Composition	Orientation Temp.	Orientation %	(cm) Tube Width	Tongue Tear MD	Tongue Tear TD	Dart Impact (ft–lbs)
23	70% SA 872, 30% PE	EPR–10%	Hot	100%	30	5.2	3.4	2.0
24	,,	EPR–10%	Cold	50%	45	6.0	5.0	1.5
25	,,	EPR–10%	Cold	100%	45	5.0	3.2	3.0
26	,,	EPR–10%	Hot	50%	45	5.6	3.9	1.5
27	,,	EPR–10%	Hot	100%	45	5.0	5.4	2.0
28	85% 8623, 10% PE, 5% EPR	EPR–10%	Cold	100%	30	0.63	0.32	2.0
29	90% 8623, 10% EPR	EPR–10%	Cold	100%	30	0.45	0.26	2.0
30	100% SA 872	EPR/PE–20%	Cold	100%	30	4.4	4.3	3.0
31	100% SA 872	EPR/PE–5%	Cold	100%	30	4.5	3.5	2.0
32	80% SA 872, 10% PE, 10% EPR	EPR/PE–10%	Cold	100%	30	3.4	6.3	3.0
33	70% NOV, 30% PE	EPR/PE–10%	Cold	100%	30	4.8	2.4	2.5
34	70% PE, 30% NOV	EPR–10%	Cold	100%	30	4.9	4.2	4.0

TABLE (Continued)

Run	Mullen Burst (psi)	Beach Puncture MD	Beach Puncture TD	Trapezoidal Tear MD	Trapezoidal Tear TD	Elmendorf Tear MD	Elmendorf Tear TD	Basis Weight (g/m²)
1	20	129	136	6.5	7.1	500–1400	500–1300	73.5
2	30	242	228	8.9	6.8	800–	500–2500	69
3	26	295	241	7.8	6.4	300–1100	300–1000	62
4	37	86	105	11.6	9.5	1200–2400	400–2000	114
5	27	103	118	11.9	7.7	400–2000	1400–3200	73.3
6	35	66	98	10.2	11.4	100–1900	1500–3200	103.7
7	30	67	98	13.6	12.9	500–1500	2400–3000	86.5
8	20	71	105	8.7	8.8	100– 800	1100–2600	77
9	19	69	120	7.8	5.0	200–2900	2200–3200	63.4
10	25	44	52	7.7	8.2	300–1100	1100–3200	77.8
11	20	86	88	9.9	8.7	900–1900	900–3200	61.1
12	21	113	103	6.3	5.4	200–1800	600–2200	54.6
13	22	70	68	7.9	5.8	310	800–2100	79
14	22	126	140	5.3	7.4	200–3000	1500–3200	70.4
15	26	56	44	9.5	8.1	300–3200	100– 300	83.7
16	29	72	64	10.7	7.8	150–2500	200–3200	71.5
17	29	83	105	9.8	8.9	700–3200	800–2000	84.5
18	23	327	278	12.5	8.4	700–3000	1800	88.4
19	20	212	187	10.4	9.0	1000–3200	900–1300	71.4
20	23	93	105	10.3	8.9	300–3200	1100–3200	74.8
21	21	400	115	8.2	7.6	300–3200	700–2100	70
22	26	55	71	11.5	9.4	600–2400	1200–2500	86.9

TABLE (Continued)

Run	Mullen Burst (psi)	Beach Puncture MD	Beach Puncture TD	Trapezoidal Tear MD	Trapezoidal Tear TD	Elmendorf Tear MD	Elmendorf Tear TD	Basis Weight (g/m^2)
23	20	110	116	8.6	10.2	200–2200	700–1600	68.8
24	26	67	149	13.2	10.1	1400–3000	800–2000	88.7
25	23	124	145	11.5	7.9	1100–2500	1000–2300	82.9
26	23	120	101	9.6	9.3	320–3200	1200–3200	83
27	21	116	113	5.7	8.9	640–3200	600–2700	76
28	20	105	50	2.4	1.8	350	96	66.3
29	15	66	73	1.8	1.5	240	96	74
30	24	99	94	9.0	7.7	800–2000	2600–3200	73.8
31	23	74	107	8.6	8.4	1400–3000	700–2700	81.7
32	25	110	117	8.4	9.4	700–2000	2400	70.4
33	23	228	194	7.0	7.9			68
34	23	336	343	8.6	6.9			71.2

WHAT I CLAIM IS:—

1. A method of producing a laminate of at least two layers, comprising the steps of rotating relative to one another in a circular coextrusion die having an exit slot at least two concentric tubular layers, each comprising a stream or an array of streams of molten polymer material, and simultaneously melt stretching each of said layers substantially in one direction, subsequently bonding the layers in the die immediately prior to their passage through the exit slot to form a laminate with the directions of melt stretching crossing each other and solidifying the laminate while maintaining the crossing melt stretched structure, the bonding in the solidified laminate being sufficiently weak to permit local delamination of the film upon tearing of the laminate.

2. A method according to claim 1, in which the opposite sides of the exit slot through which the laminate is extruded are rotated relative to one another, whereby the laminate is subjected to shear during extrusion.

3. A method according to claim 1 or claim 2, in which each of the tubular layers is

formed of a polymer-in-polymer dispersion so as to produce by the melt stretching a grain of polymer along the direction of melt stretching.

4. A method according to any of the preceding claims, in which two layers are rotated in different directions but with substantially the same angular speed.

5. A method according to any of the preceding claims, in which the generally weak bond is produced by forming the layers of polymers that adhere poorly to each other.

6. A method according to any of claims 1 to 4, in which the generally weak bond is produced by coextruding between the said layers a polymer for controlling the adhesion strength.

7. A method according to claim 6, in which the adhesion controlling polymer is extruded in stripes or is otherwise interrupted.

8. A method according to claim 6 or claim 7, in which the adhesion controlling polymer is an elastomer with a poor adhesion for the polymer material or materials which form the tubular layers.

9. A method according to any of the preceding claims, in which each layer is an array of streams and the melt stretching is performed by passing the material through a row of partitions.

10. A method according to any of claims 1 to 8, in which the melt stretching is performed by a reduction of the thickness of the molten tubular layer.

11. A method according to any of the preceding claims, in which prior to the uniting of the layers with mutually crossing directions of stretching, each layer is formed by passing two or more tubular layers of different polymer materials together through a common rotated die part and coextruding them into a common chamber of the same rotated diepart to form a composite rotated tubular layer.

12. A method according to claim 11, in which the coextrusion in the common rotated diepart takes place over a circular edge.

13. A method according to any of the preceding claims, in which the solidified laminate is biaxially stretched in the solid state in at least two separate steps each of which is essentially unidirectional.

14. A method according to claim 13, in which the stretching in the solid state is caried out substantially at room temperature.

15. A method according to claim 14, comprising several steps of substantially transverse stretching in a nip between grooved rollers with the grooves being parallel with or forming a small angle, with the machine direction.

16. A method according to claim 15, comprising longitudinally stretching the laminate after the transverse stretching.

17. A method according to claim 16, in which a substantial transverse contraction is effected during the longitudinal stretching.

18. A method according to any of the preceding claims, in which the polymer material for the tubular streams consists mainly of polyolefin.

19. A method according to claim 18, in which the polymer material for at least one of the tubular streams consists mainly of crystallisable polypropylene.

20. A method according to claim 18, in which the polymer material for at least one of the tubular streams consists mainly of high density polyethylene.

21. A method according to claim 19, in which for control of bonding strength ethylene-propylene rubber is coextruded between the tubular layers.

22. A laminate made by a method according to any preceding claim.

23. Apparatus suitable for use in a method according to claim 1 comprising a circular coextrusion die having an exit slot, means for feeding towards the slot at least two concentric tubular layers each comprising a stream or an array of streams of molten polymeric material, means for rotating the layers relative to one another in the die and for simultaneously melt stretching each layer substantially in one direction, means for co-extruding between the said layers a polymer for controlling the adhesion strength, and means for bonding the layers in the die, with their directions of melt stretching crossing each other, immediately prior to their passage from the exit slot.

24. Apparatus according to claim 23 in which the means for rotating the layers in the die relative to one another comprise means for rotating the opposite sides of the exit slot relative to one another.

25. Apparatus according to claim 23 or claim 24 in which the means for forming each of the layers comprises means for passing two or more tubular layers of different polymer materials together through a common rotated die-part and means for coextruding them into a common chamber of the same die-part.

26. Apparatus according to claim 24 substantially as herein described.

27. Apparatus according to claim 23 substantially as herein described with reference to Figure 2.

For the Applicant:—
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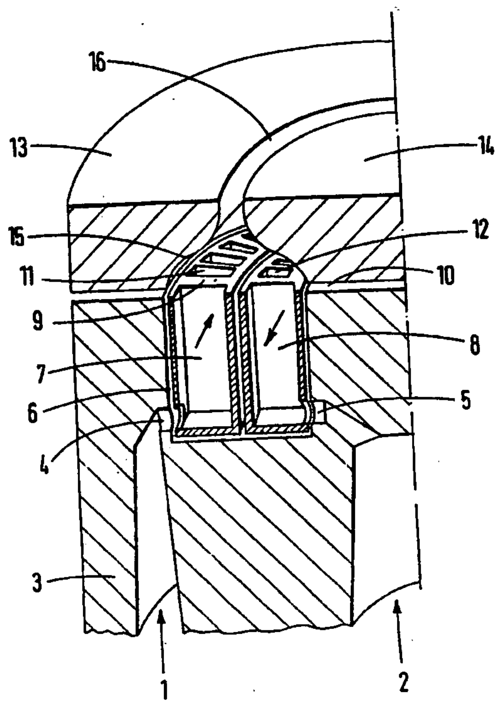
1526723 COMPLETE SPECIFICATION

4 SHEETS *This drawing is a reproduction of the Original on a reduced scale*

Sheet 1

Fig. 1



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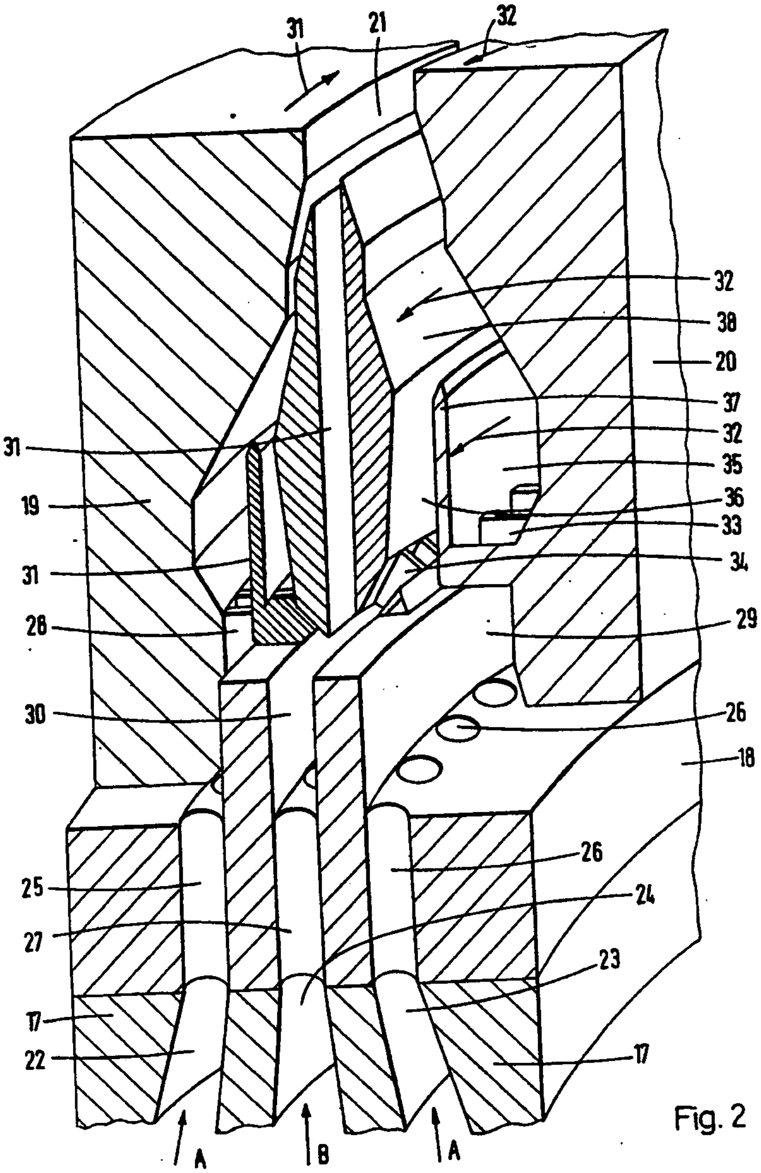

Fig. 2

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Sheet 3

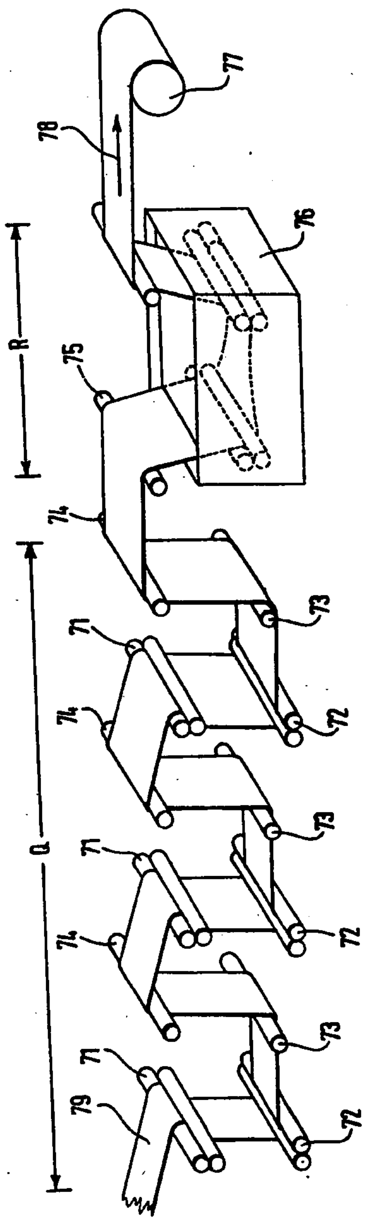

Fig. 3

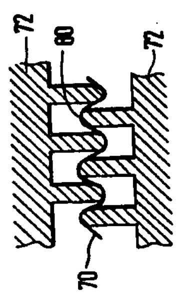

Fig. 4

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Sheet 4

Fig. 5

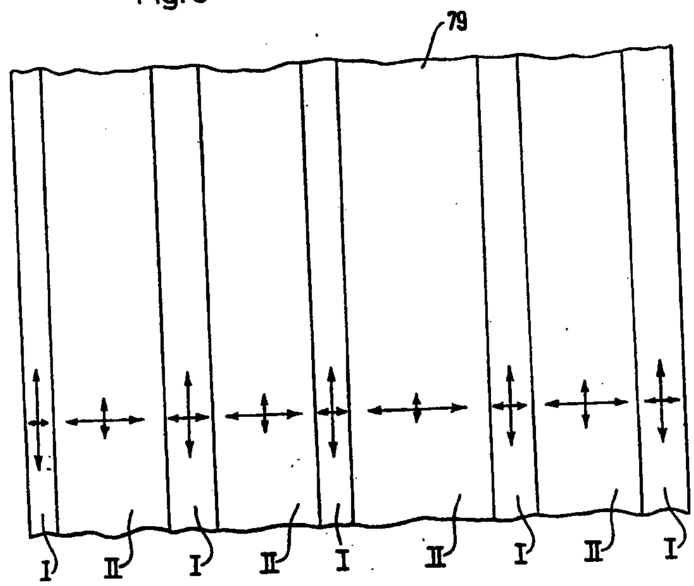

Fig. 6